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14. ABSTRACT The purpose of this project was to develop sensitive room temperature optical magnetometers based on the principles of cavity optomechanical sensing. Theoretical modelling predicts that sensitivity is possible that exceeds the current state-of-the-art in SQUID magnetometers, but with microwatts of power consumption, and without the need for cryogenics. This project was to implement magnetometers at both micro- and cm-scale with the aim of getting close to the theoretical predictions. In the first (funded) year of the project, we have achieved microscale sensitivities at the level of 200 picometers per root Hertz, a factor of two away from the best microscale room					
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## Report Title

### Achieving High Sensitivity in Cavity Optomechanical Magnetometry

#### ABSTRACT

The purpose of this project was to develop sensitive room temperature optical magnetometers based on the principles of cavity optomechanical sensing. Theoretical modelling predicts that sensitivity is possible that exceeds the current state-of-the-art in SQUID magnetometers, but with microwatts of power consumption, and without the need for cryogenics. This project was to implement magnetometers at both micro- and cm-scale with the aim of getting close to the theoretical predictions. In the first (funded) year of the project, we have achieved microscale sensitivities at the level of 200 picometers per root Hertz, a factor of two away from the best microscale room temperature magnetometers, but still two orders of magnitude away from equivalent cryogenic SQUIDS. Using the non-linear structural response we have achieved functional devices for field frequencies as low as 2 Hz. These results have been patented and submitted for publication. We have also constructed cm-scale magnetometers but have yet to fully test them. The results are in line with our milestones for the first year, with prospects for improved sensitivity in microscale devices and the full implementation of cm-scale devices planned for a second follow-on year.

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**Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:**

**(a) Papers published in peer-reviewed journals (N/A for none)**

<u>Received</u>	<u>Paper</u>
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**TOTAL:**

**Number of Papers published in peer-reviewed journals:**

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**(b) Papers published in non-peer-reviewed journals (N/A for none)**

<u>Received</u>	<u>Paper</u>
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**TOTAL:**

Number of Papers published in non peer-reviewed journals:

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(c) Presentations

Ultrasensitive Cavity Optomechanical Magnetometry  
Eoin Sheridan, Stefan Forstner, Halina Rubinszstein-Dunlop, and Warwick P. Bowen  
CLEO Europe 2013, Munich

Cavity Optomechanical Magnetometry (Invited)  
Eoin Sheridan, Stefan Forstner, Halina Rubinszstein-Dunlop, and Warwick P. Bowen  
OSA Advanced Sensors 2013, Puerto Rico

Whispering gallery mode based nanoparticle, field, and force sensing (invited)  
W. P. Bowen, U. L. Andersen, A. Doherty, S. Forstner, G. I. Harris, J. Knittel, H. Rubinszstein-Dunlop, E. Sheridan, T. Stace, J. Swaim, and M. A. Taylor  
Workshop on the Physics of Quantum electronics (PQE), 2013 Snowbird, Utah

Ultrasensitive nanoparticle, force, and field sensing with microtoroidal resonators (Invited)  
W. P. Bowen, U. L. Andersen, A. Doherty, S. Forstner, G. I. Harris, J. Knittel, H. Rubinszstein-Dunlop, E. Sheridan, T. Stace, J. Swaim1, and M. A. Taylor  
International Workshop on Microcavities and their Applications (WOMA), Shanghai, China, 2013

Number of Presentations: 4.00

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Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

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Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received              Paper

08/12/2013    1.00    S. Forstner, E. Sheridan, J. Knittel, C. L. Humphreys, G. A. Brawley, H. Rubinsztein-Dunlop, W. P. Bowen. Ultrasensitive optical magnetometry at the microscale, Manuscript under review (07 2013)

TOTAL:              1

Number of Manuscripts:

Books

Received              Paper

TOTAL:

Patents Submitted

E. Sheridan and W. P. Bowen. Magnetometer and method of fabrication. Australian Provisional Patent #2013903621 (2013).

Patents Awarded

Awards

Bowen Awarded JILA Visiting Fellowship to collaborate on cavity optomechanics in December 2014

Graduate Students

NAME	PERCENT SUPPORTED	Discipline
Changqiu Yu	0.25	
FTE Equivalent:	0.25	
Total Number:	1	

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### Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Eoin Sheridan	1.00
<b>FTE Equivalent:</b>	<b>1.00</b>
<b>Total Number:</b>	<b>1</b>

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### Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
<b>FTE Equivalent:</b>	
<b>Total Number:</b>	

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### Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Liu Qiu	0.20	Physics
William Martin	0.30	Physics
<b>FTE Equivalent:</b>	<b>0.50</b>	
<b>Total Number:</b>	<b>2</b>	

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### Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: ..... 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 1.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense ..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: ..... 0.00

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### Names of Personnel receiving masters degrees

<u>NAME</u>
<b>Total Number:</b>

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### Names of personnel receiving PhDs

<u>NAME</u>
<b>Total Number:</b>

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**Names of other research staff**

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Terry McRae	0.07
Stefan Forstner	0.18
<b>FTE Equivalent:</b>	<b>0.25</b>
<b>Total Number:</b>	<b>2</b>

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**Sub Contractors (DD882)**

**Inventions (DD882)**

**Scientific Progress**

Attached

**Technology Transfer**

## Foreword

This project aims to develop a new approach to room temperature magnetometry based on cavity optomechanical devices. Our modelling predicts that sensitivity competitive with, or surpassing, the state-of-the-art cryogenic magnetometers should be possible, but using only microwatts of power and at room temperature. This first year of the project fabricated new magnetometer designs both at microscale and at centimetre scale. With the microscale magnetometers, we have achieved a three order of magnitude improvement in sensitivity over our previous published results, and competitive with the best microscale room temperature magnetometers. While the centimetre-scale devices are yet to be fully tested. Outputs from the project include four conference presentations, three of which were invited, a provisional patent, and a submitted journal publication.

## List of Appendixes, Illustrations and Tables

Fig. 1 (a) Old cavity optomechanical magnetometer architecture with Terfenol-D affixed to the top of the toroid. (b) New cavity optomechanical magnetometer architecture with Terfenol-D affixed inside the toroid in a specially designed cavity.

Fig. 2 Schematic of experimental set-up. FPC: Fiber polarization controller.

Fig. 3 Sensitivity as a function of frequency in linear mode of operation

Fig. 4 Low frequency sensitivity achieved by nonlinear mixup as a function of signal frequency.

Fig. 5 Photograph of assembled CaF crystal resonator sensor.

Fig. 6 Photographs of magnetic shield. Left: side view. Right: top view with shielding caps from each layer removed.

## Statement of the problem studied

Magnetometers are important for diverse applications such as geological surveys, material science, medical imaging, and defence [1]. For many of these applications sensitivity is a key metric. The current state-of-the-art in ultra-sensitive magnetometry is provided by Superconducting Quantum Interference Devices (SQUIDs) and Spin Exchange Relaxation Free magnetometers (SERFs), which enable detection of pico- to femtoTesla magnetic fields. However, technical limitations constrain the breadth of applications. SQUIDs require a cryogenic environment, increasing complexity, cost, size, and power consumption. SERFs typically have sub-kHz bandwidth, are difficult to integrate, and require magnetic shielding due to their low dynamic range [2, 3]. By contrast, magnetostrictive magnetometers suffer none of these drawbacks, but typically have two to five orders of magnitude worse sensitivity.

Magnetostrictive alloys, such as Terfenol-D, physically deform in the presence of a magnetic field. This physical deformation is read-out capacitively in typical magnetostrictive magnetometers. Crucially, sensitivity is constrained by the capacitive read-out, not fundamental noise sources such as thermomechanical fluctuations. We have previously developed a new type of magnetostrictive magnetometer based on an on-chip micro-scale cavity optomechanical system [4], where

mechanical motion is strongly coupled to the optical cavity resonance frequency. Laser light is fiber-optically coupled into the cavity, with detection of the emitted field allowing ultrasensitive read-out of mechanical deformations. These magnetometers have the potential to offer comparable sensitivity to the state-of-the-art in the field, offer significant advantages in size, resolution, bandwidth, dynamic range and power consumption, and do not require cryogenic cooling. They consequently have significant potential for applications in magnetometry such as ultralow field magnetic resonance imaging, magnetoencephalography [5], uranium enrichment [6] and detection of liquid explosives [7].

#### Summary of the most important results

As outlined in our proposal, we aim to develop two types of cavity optomechanical magnetometer; a microscale toroid based on-chip magnetometer and a cm-scale crystal resonator based magnetometer. The six-monthly milestones outlined in the proposal for the first 12 months (11/12-11/13) of the project are detailed below (status indicated in parenthesis).

##### Milestones to end June 2013:

- Achieve peak sensitivity in the picoTesla range with on-chip magnetometers (achieved)
- Characterize low frequency sensitivity of on-chip magnetometers (achieved)

##### Milestones to end December 2013:

- Demonstrate nonlinear mix-up of low frequency signals with on-chip magnetometers, and characterize sensitivity (achieved)
- Demonstrate magnetic field sensing with cm-scale magnetometers (in progress)

##### *Microscale Toroidal on-chip Magnetometer:*

The sensitivity of the initial design of our cavity optomechanical magnetometer was limited by poor coupling of the magnetostrictive material (Terfenol-D) to the optical cavity (toroid) [4]. The reason for this poor coupling was that the Terfenol-D was affixed on top of the toroid (Fig. 1a) so that deformation in the presence of a magnetic field did not act directly on the torus, and poorly interacted with the most sensitive flexural mode of the toroid. Therefore, in order to improve sensitivity, a new sensor architecture was designed and fabricated. This new architecture, illustrated in Fig.1b, placed the Terfenol-D inside a cavity within the toroid so that magnetostrictive deformation could act directly on the torus. The new architecture was fabricated at UQ using successive photolithographic, wet (HF) and dry (XeF<sub>2</sub> gas) etches and laser reflow steps before the Terfenol-D material (Etrema) was micropositioned into the cavity and held in place with epoxy.



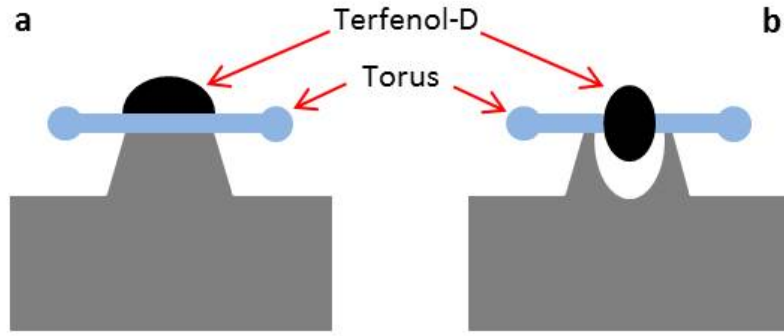


Fig. 1 (a) Old cavity optomechanical magnetometer architecture with Terfenol-D affixed to the top of the toroid. (b) New cavity optomechanical magnetometer architecture with Terfenol-D affixed inside the toroid in a specially designed cavity.

*Microscale Toroidal on-chip Magnetometer (High Frequencies):*

The final devices exhibit typical optical quality factors exceeding  $10^6$  with multiple relatively broad mechanical modes ( $Q \sim 40$ ) providing broadband mechanical response over the frequency range from 5 to 40 MHz. This allows the fundamental thermal noise limit to micromechanical magnetometry to be reached, a limit typically precluded by several orders of magnitude in magnetostrictive magnetometers that rely on electrical readout.

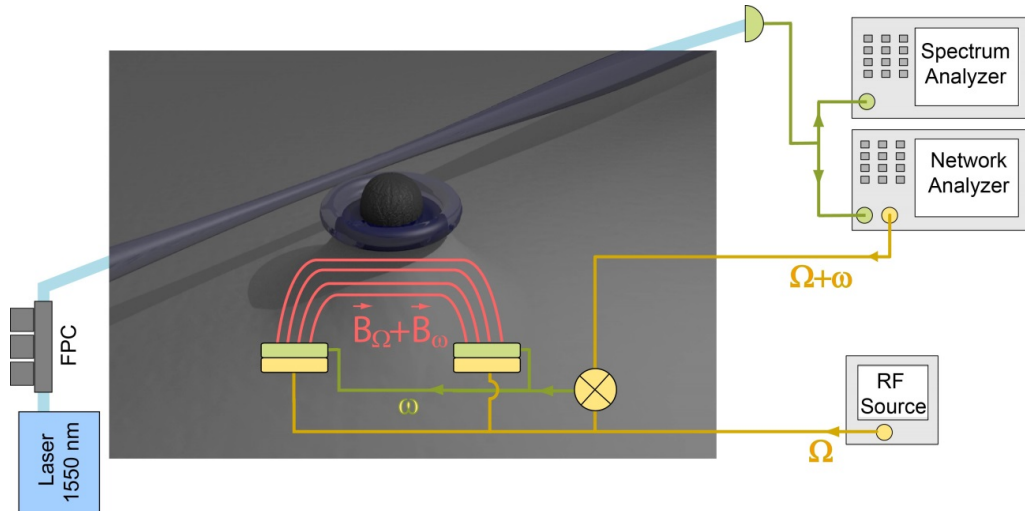


Fig. 2 Schematic of experimental set-up. FPC: Fiber polarization controller.

The sensitivity of the final magnetometer is characterized using the experimental setup shown in Fig. 2. For linear measurements of the high frequency sensitivity, the Terfenol-D is magnetized using a permanent bias magnet to maximize the linear magnetostrictive response. A pair of solenoids is then used to create a spatially uniform signal magnetic field across the device at radio frequencies. Light from a 1550 nm shot noise limited tunable fiber laser is guided through a polarization controller and evanescently coupled into the microtoroidal optical cavity via a tapered optical fiber. The laser frequency is thermally locked to the half maximum of an optical resonance. Strain in the resonator induced by the Terfenol-D shifts the optical resonance frequency, thus modulating the amplitude of the transmitted light. This transmitted field is detected on an InGaAs photodiode, with only 50  $\mu$ W

of resonant light required at the detector to achieve good signal-to-noise. Network/spectral analysis allow the magnetic field sensitivity to be determined as a function of signal frequency.

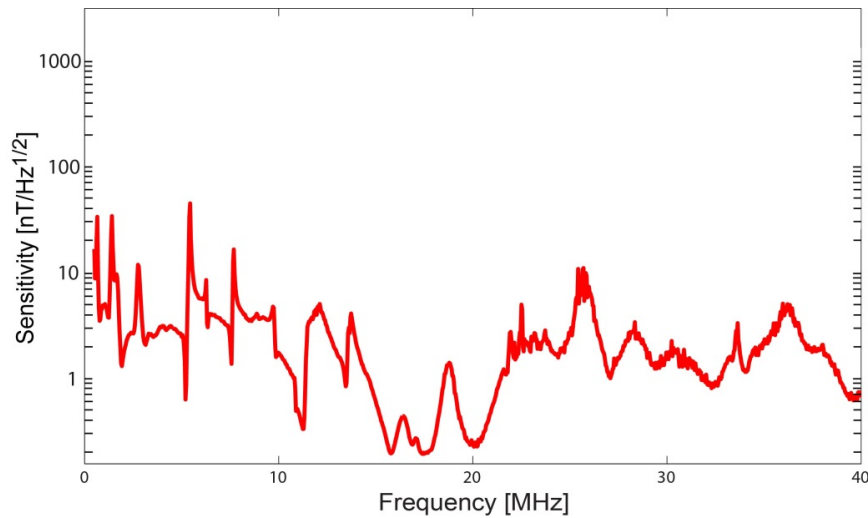


Fig.3 Sensitivity as a function of frequency in linear mode of operation.

The measured magnetic field sensitivity of the device is shown in Fig. 3 over a range from 1 to 40 MHz. As can be seen, the sensitivity exceeds  $10 \text{ nT Hz}^{1/2}$  over the majority of the range, with a peak sensitivity of  $200 \text{ pT Hz}^{1/2}$  achieved at frequencies near 20 MHz, where the dominant mechanical modes are predominantly radial in nature and thus strongly coupled to the optical resonance frequency. This represents a three orders of magnitude improvement in sensitivity on the previous design of the sensor [4]. The detection bandwidth of 1 to 40 MHz shown in Fig. 3 also represents a significant improvement on the previous sensor design [4]. The dynamic range of the sensor was tested by varying the amplitude of the signal field over several orders of magnitude. The response was linear over the range  $200 \text{ pT Hz}^{1/2}$  to  $150 \text{ } \mu\text{T Hz}^{1/2}$ , well above earth field.

#### *Microscale Toroidal on-chip Magnetometer (Low Frequencies):*

Many significant applications require sensitivity to magnetic fields in the Hz to kHz frequency regime, including magnetic anomaly detection (MAD) [8], geophysical surveys [9] and magnetoencephalography (MEG) [10]. This regime is precluded in the linear mode of operation of our device due to low frequency technical noise from vibrations, temperature fluctuations, and electronic noise. Furthermore, the mechanical resonance frequencies of micrometer-scale structures, such as microtoroids, naturally lie in the MHz-range such that the mechanical response to the magnetostrictive force is intrinsically weaker at low frequencies. However, the magnetic domain structure of magnetostrictive materials make them intrinsically highly non-linear [11]. This provides a mechanism through which to mix low frequency magnetic signals up to higher frequencies. Essentially, changes in the magnetic domain structure of the material due to low frequency fields alter the response of the material to an applied high frequency field [12]. To explore such phenomena in cavity optomechanical magnetometers, a strong RF and weak Hz/kHz magnetic field are simultaneously applied to the device and the response analysed. An RF source is used to produce an RF signal at frequency  $\Omega$ , while the network analyser produces a second RF signal at frequency  $\Omega+\omega$ , where  $\omega$  is swept in frequency over a Hz-kHz range. The signal at  $\Omega$  is split on an RF power splitter and applied to both a pair of air solenoids to generate the strong RF magnetic field, and to a mixer along with the signal at frequency  $\Omega+\omega$ , producing a low frequency signal at frequency  $\omega$ . The

low frequency signal is applied to a second set of independent air solenoids to produce the low frequency magnetic field. Any nonlinear mixing between the low and high frequency fields caused by the nonlinear magnetostrictive response can be analyzed, similarly to the case of linear operation, using spectral and network analysis.

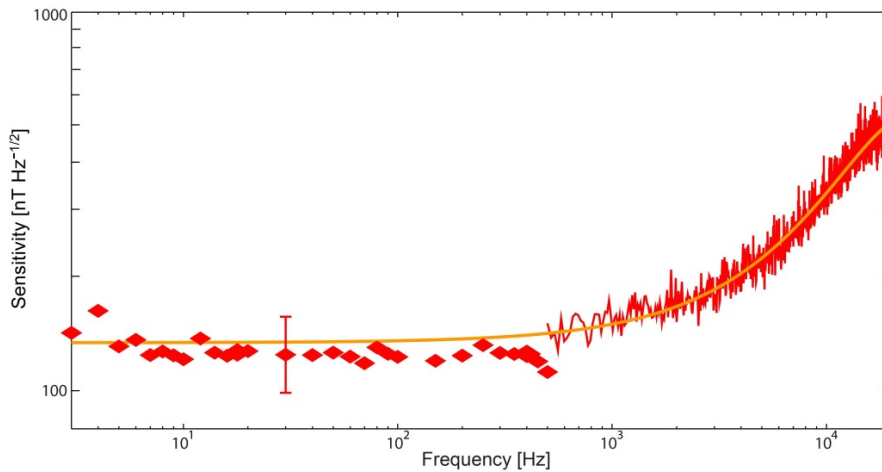


Fig. 4 Low frequency sensitivity achieved by nonlinear mixup as a function of signal frequency. At frequencies above 500 Hz the sensitivity was determined using a combination of network and spectrum analysis at 10 Hz resolution bandwidth. At lower frequencies, the network response was determined individually from each power spectrum for each data point, at a resolution bandwidth of 1 Hz. The error bar shown for a single low frequency data-point is representative of the uncertainty of all points.

The sensitivity of the nonlinear mode of operation was determined in a similar way as that for the linear mode of operation, but this time using the permanent bias magnet to maximize the nonlinear response of the device. Fig. 4 shows the achieved sensitivity at frequencies between 2 Hz and 20 kHz, with a peak sensitivity of  $150 \text{ nT Hz}^{1/2}$  over the range 2 Hz to 1 kHz. The sensitivity deteriorates at higher frequencies, probably constrained by the bandwidth of the mechanical nonlinearity. Although not shown in the figure, the sensitivity also deteriorates at frequencies below 0.5 Hz, where we observe discrete steps in the magnetometer response attributed to Barkhausen noise from random domain flips in the Terfenol-D.

#### *Crystal Resonator (cm-scale) Magnetometer:*

The principle of the CaF crystal resonator magnetometer is the same as the microtoroid based analogue. Deformation of a piece of Terfenol-D in a magnetic field is read out optically by the crystal resonator. The larger size and optical finesse of the crystal enables greater optical quality factors up to  $10^{10}$  to be reached [13], significantly greater than seen in microtoroids. It is anticipated that this will result in enhanced sensitivity, at the expense of greater sensor size, meaning that the two types of sensor will each be suited to different applications.

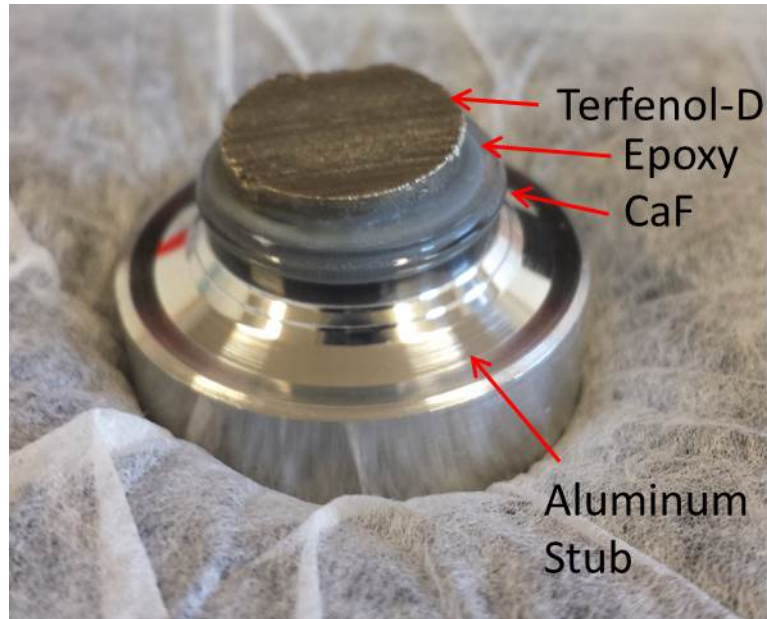


Fig. 5 Photograph of assembled CaF crystal resonator sensor.

The cm-scale magnetometers have been successfully fabricated, in a collaboration with colleagues at the Australian National University. The resonator fabrication was performed by diamond turning and polishing a CaF crystal to produce a smoothed exterior surface. To enable incorporation of Terfenol-D inside the crystal structure a diamond lathe was used to create a cylindrical hollow within the crystal. A Terfenol-D rod of diameter 12.4 mm (Etrema) was diced into 4 mm high disks using a high-speed diamond saw at UQ. The disk was fixed inside the CaF resonator with epoxy, thus completing the sensor as shown in Fig. 5.

Laser light has been coupled into the devices using a prism coupling technique, where the evanescent field of light total-internally-reflecting from a prism surface is made to overlap with the optical mode in the resonator. Through this method we have achieved 20% input coupling, and observed optical resonances with optical quality factors exceeding our design spec of  $10^9$ . The next step is to measure the response of the optical resonance frequencies to the presence of a magnetic field and calculate the sensitivity. The sensitivity should be in the range of femtotesla per root Hertz. At these magnetic field levels, background noise from the lab environment will become a critical problem. To suppress magnetic field noise we have designed and had built a six stage mu-metal magnetic shield, with design spec of background field suppression at the level of 2 femtotesla. Photographs of this shield are shown in Fig. 6.



Fig. 6 Photographs of magnetic shield. Left: side view. Right: top view with shielding caps from each layer removed.

#### *Future potential:*

The microscale magnetometers we have developed have equivalent sensitivity to the most sensitive room temperature magnetometers ever produced. This opens up possibilities for a range of applications that we would wish to pursue. Furthermore, several options exist for improved fabrication, including XeF<sub>2</sub> under-etching of the Terfenol-D to fully release it from the silicon substrate, and therefore enhance its expansion and sensitivity, and the use of different types of magnetostrictive materials. Consequently, we are confident that with some further work, cavity optomechanical magnetometers will become the leading room temperature magnetometers, at least in the laboratory, with substantial work required for commercialization; perhaps even surpassing microscale cryogenic SQUIDS.

The infrastructure is now in place in our lab to fabricate, test, and optimize cm-scale cavity optomechanical magnetometers, which have the potential to redefine the absolute state-of-the-art in magnetometer sensitivity. Such devices could have many applications ranging from MRI to MAD. We would wish to pursue both further device development and applications.

#### Bibliography

- [1] D. Robbes, *Sensors and Act. A.* 129 8693 (2006).
- [2] D. Budker, and M. Romalis, *Nat. Phys.* 3 227 (2007).
- [3] V. I. Yudin, A. V. Taichenachev, Y. O. Dudin, V. L. Velichansky, A. S. Zibrov, and S. A. Zibrov, *Phys. Rev. A* 82 033807 (2010).
- [4] S. Forstner, S. Prams, J. Knittel, E. D. van Ooijen, J. D. Swaim, G. I. Harris, A. Szorkovszky, W. P. Bowen, H. Rubinsztein-Dunlop, *Phys. Rev. Lett.* 108, 120801 (2012).
- [5] V. S. Zotev et al., *J. Mag. Res.* 194 115120 (2008).
- [6] P. L. Volegov, A. N. Matlashov, R. H. Kraus, *J. Magn. Reson.* 183 134141 (2006).
- [7] M. Espy et al., *Supercond. Sci. Technol.* 23 034023 (2010).
- [8] Zhai, J. Xing, Z. Dong, S. Li, J. & Viehland, *D. Appl. Phys. Lett.* 88, 062510 (2006).
- [9] Meyer, H. G. Stolz, R. Chwala, A. Schulz, *M. Phys. Stat. Sol.* 2 15041509 (2005).

- [10] Xia, H. Ben-Amar Baranga, A. Homan, D. & Romalis, M. V. Appl. Phys. Lett. 89 211104 (2006).
- [11] Engdahl, G. Handbook of Giant Magnetostrictive Materials (Academic Press, San Diego, 1999).
- [12] Dagenais, D. M. Bucholtz, F. & Koo, K. P. Appl. Phys. Lett. 53 1474 (1988).
- [13] A. A. Savchenkov, V. S. Ilchenko, A. B. Matsko, and L. Maleki, Phys. Rev. A 70, 051804(R) (2004).